

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER

Mo-6332/LeA 33.062

U.S. APPLICATION NO. (If known, see 37 CFR 1.5

To Be Assigned
09/831097

INTERNATIONAL APPLICATION NO.

INTERNATIONAL FILING DATE

PRIORITY DATE CLAIMED

PCT/EP99/08182 ✓

October 28, 1999 ✓

November 10, 1998 ✓

TITLE OF INVENTION THERMOPLASTIC RESINS WITH A LOW PROPORTION OF COARSE PARTICLES

APPLICANT(S) FOR DO/EO/US ZOBEL, Michael; WITTMANN, Dieter; EICHENAUER, Herbert; EITEL, Alfred; ECKEL, Thomas; KELLER, Bernd; HAUERTMANN, Hans-Bernhard and ROSE, Dorothy ✓

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
 - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ has been communicated by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☐ is attached hereto.
 - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
 - b. ☐ have been communicated by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information:

Abstract

Form PTO 1449 w/references

U.S. APPLICATION NO. (Required)
To Be AssignedINTERNATIONAL APPLICATION NO.
PCT/EP99/08182ATTORNEY'S DOCKET NUMBER
Mo-6332/LeA 33,06221. ☒ The following fees are submitted:

BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):

Neither international preliminary examination fee (37 CFR 1.482)

nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO

and International Search Report not prepared by the EPO or JPO \$1000.00

International preliminary examination fee (37 CFR 1.482) not paid to
USPTO but International Search Report prepared by the EPO or JPO \$860.00International preliminary examination fee (37 CFR 1.482) not paid to USPTO
but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00International preliminary examination fee (37 CFR 1.482) paid to USPTO
but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00International preliminary examination fee (37 CFR 1.482) paid to USPTO
and all claims satisfied provisions of PCT Article 33(1)-(4) \$100.00

ENTER APPROPRIATE BASIC FEE AMOUNT =

CALCULATIONS PTO USE ONLY

\$ 860.00

Surcharge of \$130.00 for furnishing the oath or declaration later than ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(e)).

\$ 0.00

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total claims	15 -20 =	0	x \$18.00	\$ 0.00
Independent claims	1 -3 =	0	x \$80.00	\$ 0.00

MULTIPLE DEPENDENT CLAIM(S) (if applicable)

+ \$270.00 \$ 0.00

TOTAL OF ABOVE CALCULATIONS = \$ 860.00

☐ Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above
are reduced by 1/2.

+ \$ 0.00

SUBTOTAL = \$ 860.00

Processing fee of \$130.00 for furnishing the English translation later than ☐ 20 ☐ 30
months from the earliest claimed priority date (37 CFR 1.492(f)).

\$ 0.00

TOTAL NATIONAL FEE = \$ 860.00

Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be
accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +

\$ 40.00

TOTAL FEES ENCLOSED = \$ 900.00

Amount to be
refunded: \$

charged: \$

- a. ☐ A check in the amount of \$ _____ to cover the above fees is enclosed.
- b. ☒ Please charge my Deposit Account No. 13-3848 in the amount of \$ 900.00 to cover the above fees.
A duplicate copy of this sheet is enclosed.
- c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any
overpayment to Deposit Account No. 13-3848. A duplicate copy of this sheet is enclosed.
- d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card
information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR
1.137 (a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO



00157

PATENT TRADEMARK OFFICE

SIGNATURE

Aron Preis

NAME

29,426

REGISTRATION NUMBER

09/ 831 097

JC03 Rec'd PCT/PTC

03 MAY 2001

PATENT APPLICATION
Mo-6332
LeA 33,062

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICATION OF)
) PCT/EP99/08182
MICHAEL ZOBEL ET AL)
)
SERIAL NUMBER: TO BE ASSIGNED)
)
FILED: HEREWITH)
)
TITLE: THERMOPLASTIC RESINS WITH)
)
A LOW PROPORTION OF)
)
COARSE PARTICLES)

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents

Washington, D.C. 20231

Sir:

Prior to its examination, kindly amend the English language translation of the enclosed application as follows:

"Express Mail" mailing label number ET146295674US

Date of Deposit May 3, 2001

I hereby certify that this paper or fee is being deposited with the United States Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated above and is addressed to the Assistant Commissioner of Patents and Trademarks, Washington, D.C. 20231

Donna J. Veatch

(Name of person mailing paper or fee)


Signature of person mailing paper or fee)

IN THE SPECIFICATION:

Delete the title of the application, appearing in pages 1 and 40, and insert therefor: --THERMOPLASTIC RESINS WITH A LOW PROPORTION OF COARSE PARTICLES--.

IN THE CLAIMS:

Cancel Claims 7, 14, 15 and 16.

Amend Claims 3-6 and 8-13 as follows:

3. Homo- and/or co-polymers according to Claim 1, characterised in that they contain ≤ 1000 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .

4. Homo- and/or co-polymers according to Claim 1, characterised in that they contain ≤ 500 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .

5. Homo- and/or co-polymers according to Claim 1, characterised in that they contain $\leq 10,000$ ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .

6. Homo- and/or co-polymers according to Claim 1, characterised in that they contain ≤ 5000 ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .

8. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain graft polymers of resin-forming vinyl polymers from a rubber.

9. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain thermoplastic polycarbonates and/or polyester carbonates.

10. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain polyesters.

11. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain flameproofing agents.

12. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain inorganic compounds.

13. Thermoplastic moulding compositions according to Claim 17, characterised in that they contain polyolefins.

--17. A thermoplastic molding composition comprising the homo- and/or co-polymer of Claim 1.

19. The molded article made by the method of Claim 18.--

REMARKS

The present amendment seeks to place the application in better conformance with U.S. practice. A page containing an Abstract of the Disclosure to replace the present abstract page is enclosed.

Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is titled "VERSION WITH MARKINGS TO SHOW CHANGES MADE".

Entry of the amendment is requested.

Respectfully submitted,

By



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VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Delete the title of the application, appearing in pages 1 and 40, and insert therefor: --THERMOPLASTIC RESINS WITH A LOW PROPORTION OF COARSE PARTICLES--.

IN THE CLAIMS:

Cancel Claims 7, 14, 15 and 16.

Amend Claims 3-6 and 8-13 as follows:

3. Homo- and/or co-polymers according to [either claim 1 or 2] Claim 1, characterised in that they contain ≤ 1000 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .

4. Homo- and/or co-polymers according to [any one of claims 1 to 3] Claim 1, characterised in that they contain ≤ 500 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .

5. Homo- and/or co-polymers according to [any one of claims 1 to 4] Claim 1, characterised in that they contain $\leq 10,000$ ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .

6. Homo- and/or co-polymers according to [any one of claims 1 to 5] Claim 1, characterised in that they contain ≤ 5000 ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .

8. Thermoplastic moulding compositions according to Claim [7] 17, characterised in that they contain graft polymers of resin-forming vinyl polymers from a rubber.

9. Thermoplastic moulding compositions according to [either claim 7 or claim 8] Claim 17, characterised in that they contain thermoplastic polycarbonates and/or polyester carbonates.

10. Thermoplastic moulding compositions according to [any one of claims 7 to 9] Claim 17, characterised in that they contain polyesters.

11. Thermoplastic moulding compositions according to [any one of claims 7 to 10] Claim 17, characterised in that they contain flameproofing agents.

12. Thermoplastic moulding compositions according to [any one of claims 7 to 11] Claim 17, characterised in that they contain inorganic compounds.

13. Thermoplastic moulding compositions according to [any one of claims 7 to 12] Claim 17, characterised in that they contain polyolefins.

Add the following new claims:

--17. A thermoplastic molding composition comprising the homo- and/or co-polymer of Claim 1.

18. A method of using the thermoplastic molding composition of Claim 17 comprising molding an article therefrom.

19. The molded article made by the method of Claim 18.--

THERMOPLASTIC RESINS WITH
A LOW PROPORTION OF COARSE PARTICLES

ABSTRACT OF THE DISCLOSURE

A thermoplastic molding composition having improved mechanical properties is disclosed. The composition that comprise a (co)polymer of at least one ethylenically unsaturated monomer selected from a specified group and contains less than 100 ppm of coarse portions having mean particle diameter of 200 to 500 μm , is suitable for making molded articles.

Figure 1. The effect of the initial concentration of the monomer on the polymerization of α -methylstyrene initiated by $\text{C}_6\text{H}_5\text{MgBr}$ in THF at -78°C . The concentration of the initiator was 0.01 mol/L . The concentration of the monomer was (a) 0.01 mol/L , (b) 0.02 mol/L , (c) 0.04 mol/L , (d) 0.06 mol/L , (e) 0.08 mol/L , (f) 0.10 mol/L , (g) 0.12 mol/L , (h) 0.14 mol/L , (i) 0.16 mol/L , (j) 0.18 mol/L , (k) 0.20 mol/L , (l) 0.22 mol/L , (m) 0.24 mol/L , (n) 0.26 mol/L , (o) 0.28 mol/L , (p) 0.30 mol/L , (q) 0.32 mol/L , (r) 0.34 mol/L , (s) 0.36 mol/L , (t) 0.38 mol/L , (u) 0.40 mol/L , (v) 0.42 mol/L , (w) 0.44 mol/L , (x) 0.46 mol/L , (y) 0.48 mol/L , (z) 0.50 mol/L .

Donna J. Veatch

(Name of person mailing paper or fee)

Signature of person mailing paper or fee

Thermoplastic moulding compositions having improved mechanical properties

JC03 Rec'd PCT/PTC

03 MAY 2001

The present invention relates to homo- and/or co-polymers and to thermoplastic moulding compositions having improved mechanical properties, and to the use thereof.

Thermoplastic moulding compositions, especially those containing homo- and/or co-polymers of one or more ethylenically unsaturated monomers, polycarbonates and polyesters, are known from a large number of publications. This is true especially of the use of ABS polymers. Reference is made to the following documents solely by way of example: DE-A-196 16 968, WO 97/40 092, EP-A-728 811, EP-A-315 868 (= US-A-4.937.285), EP-A-0 174 493 (US-A-4 983 658), US-A-5 030 675, JA-59 202 240, EP-A-0 363 608 (= US-A-5 204 394), EP-A-0 767 204, EP-A-0 611 798, WO 96/27 600, EP-A-0 754 531.

It is known from the prior art to filter polymer lattices in order to remove impurities or coarse portions. For example, Houben Weyl XIV/1, Makromolekulare Stoffe 1, pages 348 to 356 (Georg Thieme Verlag, Stuttgart, 1961) and DE-A-4 126 483 and US-A-4 747 959 describe the filtration of rubber lattices. Connections with the mechanical properties are not known from that prior art.

Thermoplastic moulding compositions having a particle diameter of from 0.20 to 0.35 μm are known from EP-A-0 704 488. A connection with elongation at tear, thermostability, natural shade and notched bar impact strength is not to be found in that prior art.

Accordingly, the object of the present invention is to make available homo- and/or co-polymers which are distinguished by improved mechanical properties, especially in respect of elongation at tear, thermostability, natural shade and notched bar impact strength, and by an especially consistent product quality.

That object is achieved by homo- and/or co-polymers of one or more ethylenically unsaturated monomers ("vinyl monomers") selected from the group of the mono- or poly-unsaturated olefins, such as ethylene, propylene, chloroprene, 1,3-butadiene, isopropene, vinyl acetate, styrene, α -methylstyrene, styrenes substituted at the nucleus, vinyl cyanides, such as acrylonitrile, methacrylonitrile, maleic anhydride, N-substituted maleimides, C₁-C₈-alkyl acrylates and methacrylates, such as methyl acrylate and methyl methacrylate, which contain ≤ 100 ppm of coarse portions having mean particle diameters of from 200 to 500 μm . Special preference is given to coarse portions in amounts of ≤ 50 ppm, especially ≤ 10 ppm.

The coarse portion of the homo- and/or co-polymers having mean particle diameters of from 100 to 200 μm is preferably less than 1000 ppm. Special preference is given to coarse portions of ≤ 500 ppm, especially ≤ 100 ppm. Portions of ≤ 50 ppm are very especially preferred.

In the case of mean particle diameters of from 50 to 100 μm , the coarse portion of the homo- and/or co-polymers is preferably $\leq 10,000$ ppm. In the case of those mean particle diameters, coarse portions of ≤ 5000 ppm, especially of ≤ 1000 ppm, are especially preferred. Coarse portions of ≤ 500 ppm are very especially preferred.

The mean particle diameter of the homo- and/or co-polymers used is preferably from 0.04 to 1 μm , preferably from 0.1 to 0.6 μm .

The mean particle diameter d_{50} (also called the particle size) is the diameter above and below which in each case 50 wt.% of the particles lie. It can be determined by means of ultracentrifuge measurement (W. Scholtan, H. Lange, Kolloid, Z. und Z. Polymere 250 (1972), 782-796).

According to the invention, the above-mentioned homopolymers or copolymers of ethylenically unsaturated monomers are used. Mixtures of various homo- and/or copolymers are also suitable.

5 There are suitable especially:

- rubber-free vinyl polymers (A.1),
- rubber-containing vinyl polymers, e.g. graft polymers of vinyl monomers
10 with a rubber (A.2),
- mixtures of rubber-free (A.1) and rubber-containing (A.2) vinyl polymers.

Preferred vinyl polymers A.1 are copolymers of, on the one hand, styrene, α -methylstyrene, styrene substituted at the nucleus, or mixtures thereof (A.1.1) and, on the
15 other hand, acrylonitrile, methacrylonitrile, (meth)acrylic acid C_1 - C_8 -alkyl esters, maleic anhydride, N-substituted maleimide, or mixtures thereof (A.1.2).

The copolymers can contain preferably from 50 to 98 wt.% of A.1.1 and from 50 to
20 2 wt.% of A.1.2.

Especially preferred copolymers A.1 are those of styrene, acrylonitrile and, optionally, methyl methacrylate, of α -methylstyrene, acrylonitrile and, optionally, methyl methacrylate, and of styrene, α -methylstyrene, acrylonitrile and, optionally,
25 methyl methacrylate.

The most well known are styrene-acrylonitrile copolymers, which can be prepared by radical polymerisation, especially by emulsion, suspension, solution or mass polymerisation. The copolymers A.1 preferably have molecular weights \overline{M}_w (weight
30 average, calculated by light scattering or sedimentation) of from 15,000 to 200,000.

Other especially preferred copolymers A.1 are randomly constructed copolymers of styrene and maleic anhydride, which can be prepared from the corresponding monomers, for example, by continuous mass or solution polymerisation with incomplete conversions. Their composition can be varied within wide limits. Preferably they contain from 5 to 25 wt.% of maleic anhydride units.

Instead of styrene, these polymers may also contain styrenes substituted at the nucleus, such as p-methylstyrene, vinyltoluene, 2,4-dimethylstyrene and other substituted styrenes, such as α -methylstyrene.

The rubber-containing vinyl polymers A.2 include, for example, graft (co)polymers having elastomeric properties, which are obtainable substantially from at least two of the following monomers: chloroprene, 1,3-butadiene, isopropene, styrene, α -methylstyrene, acrylonitrile, ethylene, propylene, vinyl acetate, C_1 - C_8 -alkyl acrylates and methacrylates. Such polymers are described, for example, in "Methoden der Organischen Chemie" (Houben-Weyl), Vol. 14/1, Georg Thieme-Verlag, Stuttgart, 1961, p. 393-406, and in C.B. Bucknall, "Toughened Plastics", Appl. Science Publishers, London 1977. In general, the polymers A.2 are partially crosslinked and generally have gel contents of over 20 wt.%, preferably over 40 wt.%.

Preferred rubber-like vinyl polymers A.2 are graft polymers of:

A.2.1 from 5 to 95 parts by weight, preferably from 30 to 80 parts by weight, of a mixture of

A.2.1.1 from 50 to 95 parts by weight of styrene, α -methylstyrene, styrenes substituted at the nucleus by halogen or by methyl, (meth)acrylic acid C_1 - C_8 -alkyl esters, or mixtures of these compounds, and

- 5 A.2.1.2 from 5 to 50 parts by weight of acrylonitrile, methacrylonitrile, (meth)acrylic acid C₁-C₈-alkyl esters, maleic anhydride, maleimides N-substituted by C₁-C₄-alkyl or by phenyl, or mixtures of these compounds, with
- A.2.2 from 5 to 95 parts by weight, preferably from 20 to 70 parts by weight, of rubber polymer having a glass transition temperature below -10°C.
- 10 Preferred graft polymers A.2 are, for example, polybutadienes grafted with styrene and/or acrylonitrile and/or alkyl acrylates or methacrylates, butadiene/styrene copolymers and acrylate rubbers; i.e. copolymers of the type described in DE-OS 1 694 173 (= US-A-3 564 077); polybutadienes grafted with acrylic or methacrylic acid alkyl esters, vinyl acetate, acrylonitrile, styrene and/or alkylstyrenes,
- 15 butadiene/styrene or butadiene/acrylonitrile copolymers, polyisobutene or polyisoprene, as are described, for example, in DE-OS 2 348 377 (= US-A 3 919 353).
- Especially preferred polymers A.2 are ABS polymers, as are described, for example,
- 20 in DE-OS 2 035 390 (= US-A-3 644 574) and in DE-OS 2 248 242 (= GB-B 1 409 275).
- Special preference is given according to the invention to graft rubbers having rubber contents of at least 50 wt.%, preferably at least 55 wt.%.
- 25 Especially preferred graft polymers A.2 are obtainable by graft polymerisation of
- α. from 10 to 70 wt.%, preferably from 15 to 50 wt.%, especially from 20 to 40 wt.%, based on graft polymer A.2, of acrylic acid esters or methacrylic

acid esters, or from 10 to 70 wt.%, preferably from 15 to 50 wt.%, especially from 20 to 40 wt.%, of a mixture of from 10 to 50 wt.%, preferably from 20 to 35 wt.%, based on the mixture, of acrylonitrile, acrylic acid esters or methacrylic acid esters, and from 50 to 90 wt.%, preferably from 65 to 80 wt.%, based on the mixture, of styrene or styrenes substituted at the nucleus, or a mixture thereof (as the graft overlay A.2.1) with

- 5
- β. from 30 to 90 wt.%, preferably from 50 to 85 wt.%, especially from 60 to 80 wt.%, based on graft polymer A.2, of a butadiene polymer having at least 10 50 wt.%, based on β, of butadiene radicals (as the graft base A.2.2).

In general, the gel content of the graft base β is at least 20 wt.% (measured in toluene), and the degree of grafting G is from 0.15 to 0.55.

- 15 Acrylic acid esters or methacrylic acid esters α are esters of acrylic acid or methacrylic acid and monohydric alcohols having from 1 to 8 carbon atoms. Special preference is given to methyl methacrylate, ethyl methacrylate, propyl methacrylate, n-butyl acrylate, tert-butyl acrylate and tert-butyl methacrylate.

- 20 In addition to butadiene radicals, the butadiene polymer β can contain up to 50 wt.%, based on β, of radicals of other ethylenically unsaturated monomers, such as styrene, acrylonitrile, C₁-C₄-alkyl esters of acrylic or methacrylic acid (such as methyl acrylate, ethyl acrylate, methyl methacrylate, ethyl methacrylate), vinyl esters and/or vinyl ethers). Polybutadiene is preferred.

25

It is known that in the graft polymerisation, the graft monomers are not polymerised on to the graft base completely; according to the invention, however, graft polymers A.2 include products obtained by polymerisation of the graft monomers in the presence of the graft base.

Other especially preferred polymers A.2 are graft polymers of

- 5 τ. from 20 to 90 wt.%, based on A.2, of acrylate rubber having a glass transition temperature below -20°C as the graft base A.2.2, and
- δ. from 10 to 80 wt.%, based on A.2, of at least one polymerisable, ethylenically unsaturated monomer, as the graft monomer A.2.1.

- 10 The acrylate rubbers of the polymers A.2 are preferably polymers of acrylic acid alkyl esters, optionally having up to 40 wt.%, based on τ, of other polymerisable, ethylenically unsaturated monomers. The preferred polymerisable acrylic acid esters include C₁-C₁₂-alkyl esters, preferably C₁-C₈-alkyl esters, for example methyl-, ethyl-, butyl-, n-octyl- and 2-ethyl-hexyl esters; haloalkyl esters, preferably halo-C₁-C₈-alkyl esters, such as chloroethyl acrylate, as well as mixtures of these monomers.
- 15

- For crosslinking, monomers having more than one polymerisable double bond may be copolymerised. Preferred examples of crosslinking monomers are esters of unsaturated monocarboxylic acids having from 3 to 8 carbon atoms and of
- 20 unsaturated monohydric alcohols having from 3 to 12 carbon atoms or of saturated polyols having from 2 to 4 OH groups and from 2 to 20 carbon atoms, such as, for example, ethylene glycol dimethacrylate, allyl methacrylate; polyunsaturated heterocyclic compounds, such as, for example, trivinyl and triallyl cyanurate; polyfunctional vinyl compounds, such as di- and tri-vinylbenzenes; and also triallyl
- 25 phosphate and diallyl phthalate.

Preferred crosslinking monomers are allyl methacrylate, ethylene glycol dimethacrylate, diallyl phthalate and heterocyclic compounds having at least 3 ethylenically unsaturated groups.

Especially preferred crosslinking monomers are the cyclic monomers triallyl cyanurate, triallyl isocyanurate, trivinyl cyanurate, triacryloylhexahydro-s-triazine, triallylbenzenes.

5

The amount of crosslinking monomers is preferably from 0.02 to 5 wt.%, especially from 0.05 to 2 wt.%, based on the graft base τ .

10

In the case of cyclic crosslinking monomers having at least 3 ethylenically unsaturated groups it is advantageous to restrict the amount to less than 1 wt.% of the graft base τ .

15

Preferred "other" polymerisable, ethylenically unsaturated monomers which, in addition to the acrylic acid esters, may optionally be used for the preparation of the graft base are, for example, acrylonitrile, styrene, α -methylstyrene, acrylamides, vinyl- C_1-C_6 -alkyl ethers, methyl methacrylate, butadiene. Preferred acrylate rubbers as graft base τ are emulsion polymers having a gel content of at least 60 wt.%.

20

Other suitable graft bases according to A.2.2 are silicone rubbers having graft-active sites, as are described, for example, in DE-OS 37 04 657, DE-OS 37 04 655, DE-OS 36 31 540 and DE-OS 36 31 539.

25

The gel content of the graft base A.2.2 is determined at 25°C in dimethylformamide (M. Hoffmann, H. Krömer, R. Kuhn, Polymeranalytik I und II, Georg-Thieme-Verlag, Stuttgart 1977).

The graft polymers A.2 can be prepared by known processes, such as mass, suspension, emulsion or mass-suspension processes.

According to the invention, filtration over a filter having a mean mesh size of $\leq 200 \mu\text{m}$ is preferred. Mesh sizes of $\leq 150 \mu\text{m}$ are especially preferred. Mesh sizes of $\leq 100 \mu\text{m}$ are most preferred.

5

It is also preferred that the mean mesh size should not be less than $50 \mu\text{m}$; since, if the mesh sizes are too small, there is the risk that the transverse forces will be too high. Moreover, the speed of filtration is slowed to such an extent that the outlay in terms of time becomes too high or the throughput is reduced too greatly.

10

Any known materials may be considered for the filters. These include, for example, metals or fabrics of any kind.

15

The homo- and/or co-polymers according to the invention prepared by means of the described filtration are distinguished by a significant improvement in their mechanical properties. In particular, the elongation at tear, natural shade, thermostability and notched bar impact strength are improved to a degree which was not foreseeable. Moreover, they have a surprisingly consistent product quality.

20

According to the invention, the described homo- and/or co-polymers can be partially replaced by other thermoplastic polymers. The other thermoplastic polymers are preferably selected from at least one thermoplastic polymer from the group of the polycarbonates, polyester carbonates, polyesters, preferably polyalkylene terephthalates, and conventional (co)polymers according to the above-described component A.1, but without the coarse portion of particles according to the invention.

25

The moulding compositions according to the invention may also contain other additives selected from at least one of the group of the flameproofing agents, anti-dripping agents, very finely divided inorganic compounds and filling and reinforcing materials.

5

The thermoplastic moulding compositions preferably contain

- 10
- A. from 0.5 to 60 parts by weight, preferably from 1 to 40 parts by weight, of the homo- and/or co-polymers according to the invention,
- 15
- B. from 40 to 99 parts by weight, preferably from 50 to 95 parts by weight, of polycarbonates and/or polyester carbonates,
- C. from 0 to 50 parts by weight, preferably from 1 to 30 parts by weight, especially from 2 to 25 parts by weight, of copolymers of styrene, α -methylstyrene, styrene substituted at the nucleus, or mixtures thereof, and acrylonitrile, methacrylonitrile, (meth)acrylic acid C_1-C_8 -alkyl esters, maleic anhydride, N-substituted maleimides, or mixtures thereof,
- 20
- D. from 0 to 45 parts by weight, preferably from 0 to 30 parts by weight, especially from 0 to 25 parts by weight, of polyalkylene terephthalate.

The sum of all the components of the moulding compositions according to the invention is 100.

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Some of component A may be replaced by conventional ABS polymers.

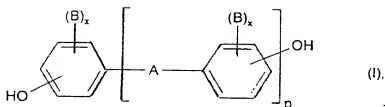
Suitable polycarbonates and/or polyester carbonates are known from the literature or can be prepared by processes which are known from the literature (for the preparation of aromatic polycarbonates see, for example, Schnell, "Chemistry and

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Physics of Polycarbonates", Interscience Publishers, 1964 and DE-AS 1 495 626, DE-OS 2 232 877, DE-OS 2 703 376, DE-OS 2 714 544, DE-OS 3 000 610, DE-OS 3 832 396; for the preparation of aromatic polyester carbonates see, for example, DE-OS 3 077 934).

The preparation of aromatic polycarbonates is effected, for example, by reaction of diphenols with carbonic acid halides, preferably phosgene, and/or with aromatic dicarboxylic acid dihalides, preferably benzenedicarboxylic acid dihalides, by the boundary surface process, optionally with the use of chain terminators, for example monophenols, and optionally with the use of branching agents having a functionality of three or more, for example triphenols or tetraphenols.

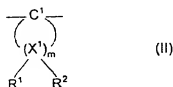
Diphenols for the preparation of the aromatic polycarbonates and/or aromatic polyester carbonates are preferably those of formula (I)

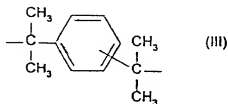


wherein

A represents a single bond, C₁-C₅-alkylene, C₂-C₅-alkylidene, C₅-C₆-cycloalkylidene, -O-, -SO-, -CO-, -S-, -SO₂-, C₆-C₁₂-arylene, to which there may be condensed further aromatic rings optionally containing heteroatoms,

or a radical of formula (II) or (III)





the substituents B each represent C₁-C₁₂-alkyl, preferably methyl, halogen, preferably chlorine and/or bromine,

the substituents x are each independently of the other 0, 1 or 2,

p represents 0 or 1, and

R¹ and R² can be selected individually for each X¹ and are each independently of the other hydrogen or C₁-C₆-alkyl, preferably hydrogen, methyl or ethyl,

X¹ represents carbon, and

m represents an integer from 4 to 7, preferably 4 or 5, with the proviso that at least one atom X¹, R¹ and R² are simultaneously alkyl.

Preferred diphenols are hydroquinone, resorcinol, dihydroxydiphenols, bis(hydroxyphenyl)-C₁-C₃-alkanes, bis(hydroxyphenyl)-C₃-C₆-cycloalkanes, bis(hydroxyphenyl) ethers, bis(hydroxyphenyl) sulfoxides, bis(hydroxyphenyl) ketones, bis(hydroxyphenyl)sulfones and α,α-bis(hydroxyphenyl)diisopropyl-benzenes and their derivatives brominated or chlorinated at the nucleus.

Especially preferred diphenols are 4,4'-dihydroxydiphenyl, bisphenol A, 2,4-bis(4-hydroxyphenyl)-2-methylbutane, 1,1-bis(4-hydroxyphenyl)-cyclohexane, 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane, 4,4'-dihydroxydiphenyl sulfide, 4,4'-dihydroxydiphenyl-sulfone and their di- and tetra-brominated or chlorinated

derivatives, such as, for example, 2,2-bis(3-chloro-4-hydroxyphenyl)-propane, 2,2-bis(3,5-dichloro-4-hydroxyphenyl)-propane or 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane.

- 5 Special preference is given to 2,2-bis(4-hydroxyphenyl)-propane (bisphenol A).

The diphenols can be used singly or in the form of any desired mixtures.

- 10 The diphenols are known from the literature or are obtainable by processes which are known from the literature.

- 15 Suitable chain terminators for the preparation of the thermoplastic, aromatic polycarbonates are, for example, phenol, p-chlorophenol, p-tert-butylphenol or 2,4,6-tribromophenol, but also long-chained alkylphenols, such as 4-(1,3-tetramethylbutyl)-phenol according to DE-OS 2 842 005 or monoalkylphenols or dialkylphenols having a total of from 8 to 20 carbon atoms in the alkyl substituents, such as 3,5-di-tert-butylphenol, p-isooctylphenol, p-tert-octylphenol, p-dodecylphenol and 2-(3,5-dimethylheptyl)-phenol and 4-(3,5-dimethylheptyl)-phenol. The amount of chain terminators to be used is generally from 0.5 mol.% to 20 10 mol.%, based on the molar sum of the diphenols used in a particular case.

- 25 The thermoplastic, aromatic polycarbonates have mean weight-average molecular weights (\overline{M}_w , measured, for example, by means of an ultracentrifuge or scattered-light measurement) of from 10,000 to 200,000, preferably from 20,000 to 80,000.

The thermoplastic, aromatic polycarbonates can be branched in a known manner, preferably by the incorporation of from 0.05 to 2 mol.%, based on the sum of the diphenols used, of compounds having a functionality of three or more, for example compounds having three or more phenolic groups.

- Both homopolycarbonates and copolycarbonates are suitable. For the preparation of copolycarbonates according to the invention, from 1 to 25 wt.%, preferably from 2.5 to 25 wt.% (based on the total amount of diphenols to be used) of polydiorganosiloxanes having hydroxy-aryloxy terminal groups may also be used.
- 5 They are known (see, for example, US patent 3 419 634) or can be prepared by processes which are known from the literature. The preparation of polydiorganosiloxane-containing copolycarbonates is described, for example, in DE-OS 3 334 782.
- 10 Preferred polycarbonates are, in addition to the homopolycarbonates of bisphenol A, the copolycarbonates of bisphenol A having up to 15 mol.%, based on the molar sum of diphenols, of diphenols other than those mentioned as being preferred or especially preferred, especially 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane.
- 15 Aromatic dicarboxylic acid dihalides for the preparation of aromatic polyester carbonates are preferably the diacid dichlorides of isophthalic acid, terephthalic acid, diphenyl ether 4,4'-dicarboxylic acid and naphthalene-2,6-dicarboxylic acid.
- 20 Mixtures of the diacid dichlorides of isophthalic acid and terephthalic acid in a ratio of from 1:20 to 20:1 are especially preferred.
- In the preparation of polyester carbonates, a carbonic acid halide, preferably phosgene, is additionally used concomitantly as bifunctional acid derivative.
- 25 As chain terminators for the preparation of the aromatic polyester carbonates, in addition to the monophenols already mentioned, their chlorocarbonic acid esters and the acid chlorides of aromatic monocarboxylic acids, which may optionally be substituted by C₁-C₂₂-alkyl groups or by halogen atoms, as well as aliphatic C₂-C₂₂-monocarboxylic acid chlorides may be considered.

The amount of chain terminators is in each case from 0.1 to 10 mol.%, based in the case of the phenolic chain terminators on moles of diphenols and in the case of monocarboxylic acid chloride chain terminators on moles of dicarboxylic acid dichlorides.

5

The aromatic polyester carbonates may also contain aromatic hydroxycarboxylic acids incorporated therein.

10

The aromatic polyester carbonates may be either linear or branched in a known manner (see in this connection likewise DE-OS 2 940 024 and DE-OS 3 007 934).

15

There may be used as branching agents, for example, carboxylic acid chlorides having a functionality of three or more, such as trimesic acid trichloride, cyanuric acid trichloride, 3,3',4,4'-benzophenone-tetracarboxylic acid tetrachloride, 1,4,5,8-naphthalenetetracarboxylic acid tetrachloride or pyromellitic acid tetrachloride, in amounts of from 0.01 to 1.0 mol.% (based on dicarboxylic acid dichlorides used), or phenols having a functionality of three or more, such as phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-hept-2-ene,

20

4,4-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-heptane, 1,3,5-tri-(4-hydroxyphenyl)-benzene, 1,1,1-tri-(4-hydroxyphenyl)-ethane, tri-(4-hydroxyphenyl)-phenylmethane, 2,2-bis[4,4-bis(4-hydroxyphenyl)-cyclohexyl]-propane, 2,4-bis(4-hydroxyphenyl-isopropyl)-phenol, tetra-(4-hydroxyphenyl)-methane,

25

2,6-bis(2-hydroxy-5-methyl-benzyl)-4-methylphenol, 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)-propane, tetra-(4-[4-hydroxyphenyl-isopropyl]-phenoxy)-methane, 1,4-bis[4,4'-dihydroxy-triphenyl-methyl]-benzene, in amounts of from 0.01 to 1.0 mol.%, based on diphenols used. Phenolic branching agents can be used initially with the diphenols, acid chloride branching agents can be introduced together with the acid dichlorides.

The content of carbonate structural units in the thermoplastic, aromatic polyester carbonates can vary as desired. The carbonate group content is preferably up to 100 mol.%, especially up to 80 mol.%, more especially up to 50 mol.%, based on the sum of ester groups and carbonate groups. Both the esters and the carbonates
5 contained in the aromatic polyester carbonates can be present in the polycondensation product in the form of blocks or in a randomly distributed manner.

The relative solution viscosity (η_{rel}) of the aromatic polycarbonates and polyester carbonates is in the range of from 1.18 to 1.4, preferably from 1.22 to 1.3 (measured
10 on solutions of 0.5 g of polycarbonate or polyester carbonate in 100 ml of methylene chloride solution at 25°C).

The thermoplastic, aromatic polycarbonates and polyester carbonates can be used alone or in any desired mixture with one another.
15

The polycarbonates may also be partially replaced by polyesters.

Preferred polyesters are polyalkylene terephthalates. These are reaction products of aromatic dicarboxylic acids (or their reactive derivatives, e.g. dimethyl esters or
20 anhydrides) and aliphatic, cycloaliphatic or arylaliphatic diols and mixtures of such reaction products.

Preferred polyalkylene terephthalates can be prepared from terephthalic acids (or their reactive derivatives) and aliphatic and cycloaliphatic diols having from 2 to 10
25 carbon atoms by known methods (Kunststoff-Handbuch, Volume VIII, p. 695 et seq., Carl Hanser Verlag, Munich 1973).

Preferred polyalkylene terephthalates contain from 80 to 100 mol.%, preferably from 90 to 100 mol.%, based on the dicarboxylic acid component, of terephthalic acid
30 radicals and from 80 to 100 mol.%, preferably from 90 to 100 mol.%, based on the

diol component, of ethylene glycol and/or 1,4-butanediol radicals. In addition to terephthalic acid radicals they also contain from 0 to 20 mol.% of radicals of other aromatic dicarboxylic acids having from 8 to 14 carbon atoms or of aliphatic dicarboxylic acids having from 4 to 12 carbon atoms, such as radicals of phthalic acid, isophthalic acid, naphthalene-2,6-dicarboxylic acid, 4,4'-diphenyldicarboxylic acid, succinic acid, adipic acid, sebacic acid, azelaic acid or cyclohexanediadicetic acid. In addition to ethylene glycol and/or 1,4-butanediol radicals, they also contain from 0 to 20 mol.% of other aliphatic diols having from 3 to 12 carbon atoms or cycloaliphatic diols having from 6 to 12 carbon atoms, for example radicals of 1,5-pentenediol, 1,6-hexanediol, 1,4-cyclohexanedimethanol, 3-methyl-1,3- and -1,6-pentenediol, 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,5-hexanediol, 1,4-di-(β -hydroxyethoxyphenyl)-propane, 2,4-dihydroxy-1,1,3,3-tetramethylcyclobutane, 2,2-bis(3- β -hydroxyethoxyphenyl)-propane and 2,2-bis(4-hydroxypropoxyphenyl)-propane (DE-OS 2 407 647, 2 407 776, 2 715 932).

The polyalkylene terephthalates can be branched by the incorporation of relatively small amounts of tri- or tetra-hydric alcohols or tri- or tetra-basic carboxylic acids, as are described in DE-OS 1 900 270 and US-A 3 692 744. Examples of preferred branching agents are trimesic acid, trimellitic acid, trimethylol-ethane and -propane and pentaerythritol. It is advisable to use no more than 1 mol.% of the branching agent, based on the acid component.

Special preference is given to polyalkylene terephthalates which have been prepared solely from terephthalic acid (or its reactive derivatives, e.g. its dialkyl esters) and ethanediol and/or 1,4-butanediol, and mixtures thereof.

Preferred polyalkylene terephthalates are also copolyesters prepared from at least two of the above-mentioned diols; especially preferred copolyesters are poly-(ethylene glycol/1,4-butanediol) terephthalates. The various diol radicals can be

present in the copolyesters in the form of blocks or in a randomly distributed manner.

5 The polyalkylene terephthalates generally have an intrinsic viscosity of from 0.4 to 1.4 dl/g, preferably from 0.5 to 1.3 dl/g, especially from 0.6 to 1.2 dl/g, in each case measured in phenol/o-dichlorobenzene (1:1 parts by weight) at 25°C.

10 The thermoplastic moulding compositions according to the invention may also contain flameproofing agents. Both halogen-containing and halogen-free compounds are suitable for that purpose. The flameproofing agents will generally be added in an amount of from 0.1 to 35 parts by weight, preferably from 0.5 to 30 parts by weight, based on the sum of components A to D.

15 Suitable halogen compounds are organic chlorine and/or bromine compounds which are stable in the preparation and processing of the moulding compositions according to the invention, so that no corrosive gases are released and the effectiveness is not impaired as a result.

20 Halogen-containing compounds are, for example,

- 21 1. Chlorinated and brominated diphenyls, such as octachlorodiphenyl, deca-chlorodiphenyl, octabromodiphenyl, decabromodiphenyl.
- 25 2. Chlorinated and brominated diphenyl ethers, such as octa- and deca-chlorodiphenyl ether and octa- and deca-bromodiphenyl ether.
3. Chlorinated and brominated phthalic anhydride and its derivatives, such as phthalimides and bisphthalimides, e.g. tetrachloro- and tetrabromo-phthalic anhydride, tetrachloro- and tetrabromo-phthalimide, N,N'-ethylene-bis-

tetrachloro- and N,N'-ethylene-bis-tetrabromo-phthalimide, N-methyltetrachloro- and N-methyltetrabromo-phthalimide.

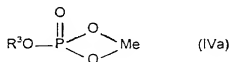
- 5 4. Chlorinated and brominated bisphenols, such as 2,2-bis(3,5-dichloro-4-hydroxyphenyl)-propane and 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane.
- 10 5. 2,2-Bis(3,5-dichloro-4-hydroxyphenyl)-propane oligocarbonate and 2,2-bis(3,5-dibromo-4-hydroxyphenyl)-propane oligocarbonate having a mean degree of polycondensation of from 2 to 20.

Bromine compounds are preferred over chlorine compounds, and halogen-free compounds are preferred over these.

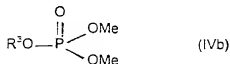
- 15 There are preferably suitable as flameproofing agents any phosphorus compounds usually used for that purpose, especially phosphine oxides and derivatives of acids of phosphorus and salts of acids and acid derivatives of phosphorus.

- 20 Derivatives (e.g. esters) of acids of phosphorus and their salts are preferred, wherein acids of phosphorus include phosphoric acid, phosphonic acid, phosphinic acid, phosphorous acid, in each case also in dehydrated form, salts are preferably alkali, alkaline earth metal and ammonium salts of these acids, and their derivatives (for example partially esterified acids) are also included.

- 25 There are suitable as phosphorus compounds, for example, metal compounds of monoesters of the phosphoric acid of formulae (IVa) and (IVb)

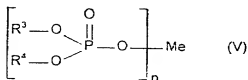


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or metal compounds of diesters of the phosphoric acid according to formula (V)

10



wherein

15

R^3 and R^4 are each independently of the other optionally halogenated C_1 - C_{24} -alkyl; C_5 - C_6 -cycloalkyl, C_6 - C_{20} -aryl or C_7 - C_{12} -aralkyl, each of which is optionally substituted by halogen and/or by C_1 - C_{10} -alkyl; or, in the case of formula (V), R^3 and R^4 together form an alkyl chain,

20

Me represents a metal selected from main groups 1 and 3 and sub-groups VIII, IB and IIB of the periodic system,

and

25

n is determined by the valency of the metal ion.

R^3 and R^4 are each independently of the other preferably optionally halogenated (preferably by chlorine and/or by bromine) C_1 - C_{15} -alkyl, especially C_1 - C_{10} -alkyl; cyclopentyl, cyclohexyl, phenyl, naphthyl, phenyl- C_1 - C_4 -alkyl (such as benzyl), each

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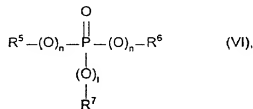
of which is optionally substituted by halogen (preferably chlorine and/or bromine) and/or by C₁-C₆-alkyl, especially C₁-C₄-alkyl, more especially methyl, ethyl, n-propyl, isopropyl.

- 5 Metals of main groups 2 and 3 and of sub-group II are preferred as the metals Me.

Me is especially preferably Mg, Ca, Ba, B, Al or Zn.

- 10 For the preparation of the metal compounds of phosphoric acid esters according to the invention there are suitable processes known from the literature, such as, for example, the transesterification process starting from triesters of phosphoric acid, or the acid halide process starting from phosphoryl chloride (EP-A-0 801 116; J. Org. Chem. 1978, Vol. 43, No. 1, p. 24-31).

- 15 Also suitable as flameproofing agents are phosphorus compounds of formula (VI)



20

wherein

- 25 R⁵, R⁶ and R⁷ are each independently of the others an optionally halogenated C₁-C₈-alkyl or an optionally halogenated and/or alkylated C₅- or C₆-cycloalkyl or an optionally halogenated and/or alkylated and/or aralkylated C₆-C₃₀aryl, and

"n" and "l" are each independently of the other 0 or 1.

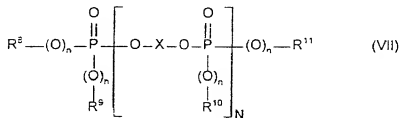
These phosphorus compounds are generally known (see, for example, Ullmann, Enzyklopädie der technischen Chemie, Volume 18, pages 301 et seq., 1979). The aralkylated phosphorus compounds are described, for example, in DE-OS 38 24 356.

- 5 Optionally halogenated C₁-C₈-alkyl radicals according to (VI) may be mono- or poly-halogenated and linear or branched. Examples of alkyl radicals are chloroethyl, 2-chloropropyl, 2,3-dibromopropyl, butyl, methyl or octyl.
- 10 Optionally halogenated and/or alkylated C₅- or C₆-cycloalkyls according to (VI) are optionally mono- to poly-halogenated and/or -alkylated C₅- or C₆-cycloalkyls, that is to say, for example, cyclopentyl, cyclohexyl, 3,3,5-trimethylcyclohexyl and fully chlorinated cyclohexyl.
- 15 Optionally halogenated and/or alkylated and/or aralkylated C₆-C₃₀-aryl radicals according to (VI) are optionally mono- or poly-nuclear, mono- or poly-halogenated and/or alkylated and/or aralkylated, for example chlorophenyl, bromophenyl, pentachlorophenyl, pentabromophenyl, phenyl, cresyl, isopropylphenyl, benzyl-substituted phenyl and naphthyl.
- 20 R⁵, R⁶ and R⁷ are each independently of the others preferably methyl, ethyl, butyl, octyl, phenyl, cresyl, cumyl or naphthyl. Especially preferably, R⁵, R⁶ and R⁷ are each independently of the others methyl, ethyl, butyl, optionally methyl- and/or ethyl-substituted phenyl.
- 25 Phosphorus compounds according to formula (VI) which can be used according to the invention are, for example, tributyl phosphate, tris(2-chloroethyl) phosphate, tris(2,3-dibromopropyl) phosphate, triphenyl phosphate, tricresyl phosphate, diphenylcresyl phosphate, diphenyloctyl phosphate, diphenyl-2-ethylcresyl phosphate, tri-(isopropylphenyl) phosphate, tris(p-benzylphenyl) phosphate,

triphenylphosphine oxide, methylphosphonic acid dimethyl esters, methanephosphonic acid diphenyl esters and phenylphosphonic acid diethyl esters.

Suitable flameproofing agents are also dimeric and oligomeric phosphates, as described, for example, in EP-A-0 363 608.

The moulding compositions according to the invention may contain as flameproofing agents phosphorus compounds according to formula (VII)



In the formula, R^8 , R^9 , R^{10} and R^{11} are each independently of the others C_1 - C_8 -alkyl, C_5 - C_6 -cycloalkyl, C_6 - C_{20} -aryl or C_7 - C_{12} -aralkyl, each of which is optionally halogenated.

R^8 , R^9 , R^{10} and R^{11} are each independently of the others preferably C_1 - C_4 -alkyl, phenyl, naphthyl or phenyl- C_1 - C_4 -alkyl. The aromatic groups R^8 , R^9 , R^{10} and R^{11} may in turn be substituted by halogen and/or alkyl groups, preferably chlorine, bromine and/or C_1 - C_4 -alkyl. Especially preferred aryl radicals are cresyl, phenyl, xylenyl, propylphenyl or butylphenyl and the corresponding brominated and chlorinated derivatives thereof.

X in formula (VII) represents a mono- or poly-nuclear aromatic radical having from 6 to 30 carbon atoms. It is derived preferably from diphenols of formula (I). Special preference is given to diphenylphenol, bisphenol A, resorcinol or hydroquinone or their chlorinated or brominated derivatives.

The substituents n in formula (VII) may, independently of one another, be 0 or 1; n is preferably 1.

N represents values of from 0 to 30, preferably an average value of from 0.3 to 20, especially from 0.5 to 10, more especially from 0.5 to 6.

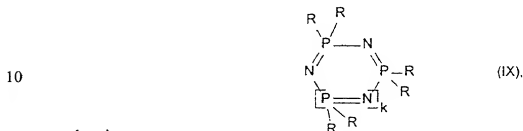
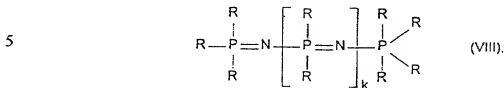
It is also possible to use mixtures of from 10 to 90 wt.%, preferably from 12 to 40 wt.%, of at least one monophosphorus compound of formula (VI) and at least one oligomeric phosphorus compound or a mixture of oligomeric phosphorus compounds as described in EP-A-363 608 as well as phosphorus compounds according to formula (VII) in amounts of from 10 to 90 wt.%, preferably from 60 to 88 wt.%, based on the total amount of phosphorus compounds.

Monophosphorus compounds of formula (VI) are especially tributyl phosphate, tris(2-chloroethyl) phosphate, tris(2,3-dibromopropyl) phosphate, triphenyl phosphate, tricresyl phosphate, diphenylcresyl phosphate, diphenyloctyl phosphate, diphenyl-2-ethylcresyl phosphate, tri-(isopropylphenyl) phosphate, halo-substituted aryl phosphates, methylphosphonic acid dimethyl esters, methylphosphonic acid diphenyl esters, phenylphosphonic acid diethyl esters, triphenylphosphine oxide or tricresylphosphine oxide.

The mixtures of monomeric and oligomeric phosphorus compounds of formula (VII) have average N values of from 0.3 to 20, preferably from 0.5 to 10, especially from 0.5 to 6.

The mentioned phosphorus compounds are known (see, for example, EP-A-363 608, EP-A-640 655) or can be prepared by known methods in an analogous manner (e.g. Ullmanns Enzyklopädie der technischen Chemie, Vol. 18, p. 301 et seq., 1979; Houben-Weyl, Methoden der organischen Chemie, Vol. 12/1, p. 43; Beilstein Vol. 6, p. 177).

The phosphorus compounds which can be used according to the invention also include linear phosphazenes according to formula (VIII) and cyclic phosphazenes according to formula (IX)



wherein

the substituents R are in each case identical or different and represent amino; C₁- to C₆-alkyl or C₁-C₈-alkoxy, each of which is optionally halogenated, preferably halogenated by fluorine; C₅- to C₆-cycloalkyl, C₆- to C₂₀-aryl, preferably phenyl or naphthyl, C₆-C₂₀-aryloxy, preferably phenoxy, naphthyloxy, or C₇-C₁₂-aralkyl, preferably phenyl-C₁-C₄-alkyl, each of which is optionally substituted by alkyl, preferably C₁-C₄-alkyl, and/or by halogen, preferably chlorine, bromine,

k represents 0 or a number from 1 to 15, preferably a number from 1 to 10.

There may be mentioned by way of examples:
propoxyphosphazene, phenoxyphosphazene, methylphenoxyphosphazene, aminophosphazene and fluoroalkylphosphazene.

Phenoxyphosphazene is preferred.

The phosphazenes can be used alone or in the form of a mixture. The radicals R can always be identical, or two or more radicals in formulae (VIII) and (IX) can be different.

- 5 The phosphazenes and their preparation are described, for example, in EP-A-728 811, DE-A-1 961 668 and WO 97/40 092.

10 The moulding compositions according to the invention may also contain preferably from 0.05 to 5 parts by weight, especially from 0.1 to 1 part by weight, more especially from 0.1 to 0.5 part by weight, based on the total composition, of fluorinated polyolefins. Suitable fluorinated polyolefins have a high molecular weight and have glass transition temperatures of over -30°C , generally over 100°C . Their fluorine contents are preferably from 65 to 76 wt.%, especially from 70 to 76 wt.%. Their mean particle diameters d_{50} are generally from 0.05 to $1000\text{ }\mu\text{m}$,
15 preferably from 0.08 to $20\text{ }\mu\text{m}$. In general, the fluorinated polyolefins E have a density of from 1.2 to 2.3 g/cm^3 .

20 Preferred fluorinated polyolefins are polytetrafluoroethylene, polyvinylidene fluoride, tetrafluoroethylene/hexafluoropropylene copolymers and ethylene/tetrafluoroethylene copolymers.

25 The fluorinated polyolefins are known (see "Vinyl and Related Polymers" by Schildknecht, John Wiley & Sons, Inc., New York, 1962, pages 484 to 494; "Fluoropolymers" by Wall, Wiley-Interscience, John Wiley & Sons, Inc., New York, Volume 13, 1970, pages 623 to 654; "Modern Plastics Encyclopedia", 1970 to 1971, Volume 47, No. 10A, October 1970, McGraw-Hill, Inc., New York, pages 134 and 774; "Modern Plastics Encyclopedia", 1975 to 1976, October 1975, Volume 52, No. 10A, McGraw-Hill, Inc., New York, pages 27, 28 and 472, and US-A 3 671 487, 3 723 373 and 3 838 092).

- They can be prepared by known processes, for example by polymerisation of tetrafluoroethylene in an aqueous medium with a catalyst that forms free radicals, for example sodium, potassium or ammonium peroxydisulfate, at pressures of from 7 to 71 kg/cm² and at temperatures of from 0 to 200°C, preferably at temperatures of from 20 to 100°C. (For further details see, for example, US patent 2 393 967.) Depending on the form in which they are used, the density of these materials may be from 1.2 to 2.3 g/cm³, and the mean particle diameter may be from 0.05 to 1000 µm.
- 10 Preferred fluorinated polyolefins are tetrafluoroethylene polymers. They have mean particle diameters of from 0.05 to 20 µm, preferably from 0.08 to 10 µm, and a density of from 1.2 to 1.9 g/cm³ and are preferably used in the form of a coagulated mixture of emulsions of the tetrafluoroethylene polymers E with emulsions of the graft polymers C.
- 15 Suitable fluorinated polyolefins which can be used in powder form are tetrafluoroethylene polymers having mean particle diameters of from 100 to 1000 µm and densities of from 2.0 g/cm³ to 2.3 g/cm³.
- 20 The thermoplastic moulding compositions according to the invention may also contain very finely divided inorganic compounds. The moulding compositions according to the invention preferably contain from 0.1 to 50 parts by weight, preferably from 0.1 to 10 parts by weight, based on the total amount. They may preferably consist of compounds of one or more metals of main groups 1 to 5 or sub-
- 25 groups 1 to 8 of the periodic system, preferably main groups 2 to 5 or sub-groups 4 to 8, especially main groups 3 to 5 or sub-groups 4 to 8, with at least one element selected from the group consisting of oxygen, sulfur, boron, phosphorus, carbon, nitrogen, hydrogen and silicon.

Preferred compounds are, for example, oxides, hydroxides, water-containing oxides, sulfates, sulfites, sulfides, carbonates, carbides, nitrates, nitrites, nitrides, borates, silicates, phosphates, hydrides, phosphites or phosphonates.

- 5 Preferred very finely divided inorganic compounds are, for example, TiN, TiO₂, SnO₂, WC, ZnO, Al₂O₃, AlO(OH), ZrO₂, Sb₂O₃, SiO₂, iron oxides, Na₂SO₄, Si, BaSO₄, vanadium oxides, zinc borate, silicates such as Al silicates, Mg silicates, one-, two-, three-dimensional silicates, mixtures and doped compounds can likewise be used. Furthermore, these nano-scale particles can be surface-modified by organic
- 10 molecules in order to achieve better tolerability with the polymers. In that manner, hydrophobic or hydrophilic surfaces can be produced.

The average particle diameters are less than or equal to 200 nm, preferably less than or equal to 150 nm, especially from 1 to 100 nm.

- 15 Particle size and particle diameter always refer to the mean particle diameter d_{50} , determined by ultracentrifuge measurements according to W. Scholtan *et al.*, Kolloid-Z. und Z. Polymere 250 (1972), p. 782 to 796.

- 20 The inorganic compounds may be in the form of powders, pastes, sols, dispersions or suspensions. Powders can be obtained from dispersions, sols or suspensions by precipitation.

- The powders can be incorporated into the thermoplastic plastics by conventional
- 25 processes, for example by direct kneading or extrusion of the constituents of the moulding composition and the very finely divided inorganic powders. Preferred processes are the preparation of a master batch, for example in flameproofing additives, other additives, monomers, solvents, the co-precipitation of dispersions of the above-described components of the thermoplastic moulding compositions

according to the invention with dispersions, suspensions, pastes or sols of the very finely divided inorganic materials.

- 5 The moulding compositions according to the invention may also contain conventional additives, such as lubricating and mould release agents, nucleation agents, antistatics, stabilisers, filling and reinforcing materials as well as colourings and pigments.

- 10 The filled or reinforced moulding compositions can contain up to 60 wt.%, preferably from 10 to 40 wt.%, based on the filled or reinforced moulding composition, of filling and/or reinforcing materials. Preferred reinforcing materials are glass fibres. Preferred filling materials, which may also have a reinforcing effect, are glass beads, mica, silicates, quartz, talcum, titanium dioxide, wollastonite.

- 15 The moulding compositions according to the invention can be prepared by mixing the constituents in a known manner and melt-compounding or melt-extruding the mixture at elevated temperatures, preferably at from 200 to 350°C, in conventional devices, such as kneaders, extruders or double-shaft screws. The constituents can be mixed in succession or simultaneously. In special cases it may be advantageous to
20 prepare pre-mixtures from the low-molecular-weight additives and the magnesium aluminium silicates.

- The thermoplastic moulding compositions according to the invention are suitable on account of their very good mechanical properties for the production of moulded
25 bodies of any kind, especially those having increased requirements as regards fracture resistance.

- The moulding compositions of the present invention can be used for the production of moulded bodies of any kind. In particular, moulded bodies can be produced by
30 injection moulding. Examples of moulded bodies which can be produced are: casing

- parts of any kind, e.g. for domestic appliances, such as juice extractors, coffee machines, mixers, for office equipment, such as monitors, printers, copiers, or covering plates for the construction sector and parts for the motor sector. In addition, they can be used in the field of electrical engineering, because they have very good electrical properties.
- 5

The moulding compositions according to the invention can also be used, for example, for producing the following moulded bodies or mouldings:

- 10
1. parts for finishing the interior of track vehicles (FR)
 2. hub caps
 3. casings for electrical appliances containing small transformers
 4. casings for devices for distributing and transmitting information
 5. casings and coverings for medical purposes
- 15
6. massage devices and casings therefor
 7. toy vehicles for children
 8. flat prefabricated wall panels
 9. casings for security devices
- 20
10. rear spoilers
 11. heat-insulated containers
 12. device for keeping or caring for small animals
 13. mouldings for sanitary and bathroom fittings
 14. grids for covering ventilation openings
 15. mouldings for garden and tool sheds
- 25
16. casings for gardening tools

Another form of processing is the production of moulded bodies by deep drawing from previously prepared plates or films.

The invention is described in greater detail below with reference to the Examples:

Examples

5 **Component A**

A1 (comparison)

10 Graft polymer of 40 parts by weight of a copolymer of styrene and acrylonitrile in a ratio of 73:27 with 60 parts by weight of particulate crosslinked polybutadiene rubber (mean particle diameter $d_{50} = 0.3 \mu\text{m}$), prepared by emulsion polymerisation.

15 The coarse portion having a particle diameter of from 200 to 500 μm is 3800 ppm (determined by wet screening with subsequent gravimetric quantitative determination of the coarse portion).

A2 (comparison)

20 Graft polymer of 40 parts by weight of a copolymer of styrene and acrylonitrile in a ratio of 73:27 with 60 parts by weight of particulate crosslinked polybutadiene rubber (mean particle diameter $d_{50} = 0.35 \mu\text{m}$), prepared by emulsion polymerisation.

25 The coarse portion having a particle diameter of from 200 to 500 μm is 4100 ppm (determined by wet screening with subsequent gravimetric quantitative determination of the coarse portion).

A3

5 Graft polymer according to the invention of 40 parts by weight of a copolymer of styrene and acrylonitrile in a ratio of 73:27 with 60 parts by weight of particulate crosslinked polybutadiene rubber (mean particle diameter $d_{50} = 0.3 \mu\text{m}$), prepared by emulsion polymerisation.

10 The coarse portion having a particle diameter of from 200 to 500 μm of the graft polymer according to the invention is less than 50 ppm (determined by wet screening with subsequent gravimetric quantitative determination of the coarse portion).

A4

15 Graft polymer according to the invention of 40 parts by weight of a copolymer of styrene and acrylonitrile in a ratio of 73:27 with 60 parts by weight of particulate crosslinked polybutadiene rubber (mean particle diameter $d_{50} = 0.35 \mu\text{m}$), prepared by emulsion polymerisation.

20 The coarse portion having a particle diameter of from 200 to 500 μm of the graft polymer according to the invention is less than 50 ppm (determined by wet screening with subsequent gravimetric quantitative determination of the coarse portion).

25

Component B

Linear polycarbonate based on bisphenol A having a relative solution viscosity of 1.252 measured in CH_2Cl_2 as solvent at 25°C and in a concentration of 0.5 g/100 ml.

Component C

- 5 Styrene/acrylonitrile copolymer having a styrene/acrylonitrile ratio of 72:28 and an intrinsic viscosity of 0.55 dl/g (measured in dimethylformamide at 20°C).

Preparation and testing of the moulding compositions according to the invention

- 10 Components A-C are mixed using a 3 litre kneader. The moulded bodies are produced using an Arburg 270 E injection moulding machine at 260°C.

The notched bar impact strength is determined according to method ISO 1801 A using rods measuring 80 x 10 x 4 mm at room temperature.

- 15 The elongation at tear DR is determined within the framework of the determination of the modulus of elasticity in tension according to method ISO 527 using F3 shoulder rods.

- 20 The yellowness index is determined in accordance with ASTM D 1925.

- 25 In the case of the thermostability, specimen plates (60 x 40 x 2 mm) are injection-moulded at a tool temperature of 80°C and a composition temperature of from 260 to 300°C. The composition temperature is reached in 10°C steps; the dwell time of the composition in the cylinder is 6 minutes.

A visual assessment of the surface is carried out according to the following criteria:

	Symbol	Assessment
	0	not tested
5	1	i.o., smooth surface, no cracks
	2	traces of disintegration, smooth surface, small, just visible cracks
	3	minimal disintegration, smooth surface, up to 5 blistered cracks
	4	slight disintegration, smooth surface, from 5 to 10 blistered cracks
	5	moderate disintegration, cracked blistered surface, approximately half
10		still smooth
	6	severe disintegration, cracked surface, still smooth areas visible
	7	very severe disintegration, foam-like condition
	8	foam formation, sputters from nozzle, cannot be injection-moulded

Table 1: Composition and properties of the polycarbonate-ABS moulding compositions

Example	1 (comparison)	2	3 (comparison)	4
Components parts by weight				
A1	24.0	-	-	-
A2	-	-	24.0	-
A3	-	24.0	-	-
A4	-	-	-	24.0
B	43.0	43.0	43.0	43.0
C	33.0	33.0	33.0	33.0
Properties				
a_k (ISO 1801 A) kJ/m ²	81.1	90.0	72.1	92.8
DR (ISO 527) %	35.5	80.2	15.8	79.5
Y1			39.40	38.84
Thermostability 260°C			2	1
Thermostability 270°C			2	2
Thermostability 280°C			3	2
Thermostability 290°C			4	4
Thermostability 300°C			6	5

Table 2: Composition and properties of the ABS moulding compositions

Example	1	2 (comparison)	3	4 (comparison)
Components parts by weight				
A1	-	40	-	-
A2	-	-	-	40
A3	40	-	-	-
A4	-	-	40	
C	60	60	60	60
Properties				
a_k (ISO 1801 A) kJ/m ²	19.0	14.6	21.1	19.5
a_k (ISO 1801 A) kJ/m ²	110	79	155	84
DR (ISO 527) %	17.4	8.6	9.6	6.7
YI	37.3	38.4	35.6	38.0

Patent claims

1. Homo- and/or co-polymers of one or more ethylenically unsaturated monomers selected from at least one from the group of the mono- or poly-unsaturated olefins, vinyl acetate, styrene, α -methylstyrene, styrenes substituted at the nucleus, vinyl cyanides, maleic anhydride, N-substituted maleimides, chloroprene, C_1 - C_8 -alkyl acrylates and methacrylates, which contain less than 100 ppm of coarse portions having a mean particle diameter of from 200 to 500 μm .
2. Homo- and/or co-polymers according to claim 1, characterised in that they contain ≤ 50 ppm of coarse portions having a mean particle diameter of from 200 to 500 μm .
3. Homo- and/or co-polymers according to either claim 1 or claim 2, characterised in that they contain ≤ 1000 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .
4. Homo- and/or co-polymers according to any one of claims 1 to 3, characterised in that they contain ≤ 500 ppm of coarse portions having a mean particle diameter of from 100 to 200 μm .
5. Homo- and/or co-polymers according to any one of claims 1 to 4, characterised in that they contain $\leq 10,000$ ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .
6. Homo- and/or co-polymers according to any one of claims 1 to 5, characterised in that they contain ≤ 5000 ppm of coarse portions having a mean particle diameter of from 50 to 100 μm .

7. Thermoplastic moulding compositions, characterised in that they contain homo- and co-polymers according to any one of claims 1 to 6.
- 5 8. Thermoplastic moulding compositions according to claim 7, characterised in that they contain graft polymers of resin-forming vinyl polymers from a rubber.
- 10 9. Thermoplastic moulding compositions according to either claim 7 or claim 8, characterised in that they contain thermoplastic polycarbonates and/or polyester carbonates.
- 15 10. Thermoplastic moulding compositions according to any one of claims 7 to 9, characterised in that they contain polyesters.
11. Thermoplastic moulding compositions according to any one of claims 7 to 10, characterised in that they contain flameproofing agents.
- 20 12. Thermoplastic moulding compositions according to any one of claims 7 to 11, characterised in that they contain inorganic compounds.
13. Thermoplastic moulding compositions according to any one of claims 7 to 12, characterised in that they contain polyolefins.
- 25 14. Use of the homo- and/or co-polymers according to any one of claims 1 to 6 for the preparation of moulding compositions.
15. Use of the moulding compositions according to any one of claims 7 to 13 for the production of moulded bodies.

16. Moulded bodies obtainable from moulding compositions according to any one of claims 7 to 13.

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Thermoplastic moulding compositions having improved mechanical properties

A b s t r a c t

The present invention relates to homo- and/or co-polymers that contain ≤ 100 ppm of coarse portions having a particle diameter of from 200 to 500 μm , to thermoplastic moulding compositions containing these homo- and/or co-polymers, and to the use of the homo- and/or co-polymers and of the moulding compositions for the production of moulded bodies, and to the moulded bodies themselves.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

"THERMOPLASTIC RESINS WITH A LOW PROPORTION OF COARSE PARTICLES"

the specification of which is attached hereto,

or was filed on **October 28, 1999** ✓

as a PCT Application Serial No. **PCT/EP99/08182** ✓

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims.

I acknowledge the duty to disclose information which is material to the patentability of this application in accordance with Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

198 51 676.2 ✓
(Number)

Germany ✓
(Country)

November 10, 1998 ✓
(Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose the material information as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)

(Application Serial No.)	(Filing Date)	(Status)
		(patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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